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TRANSURANIC CONCENTRATIONS IN REEF AND PELAGIC FISH FROM THE MARSHALL ISLANDS

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ABSTRACT

Concentrations of $^{239+240}Pu$ are reported in tissues of several species of reef and pelagic fish caught at 14 different atolls in the northern Marshall Islands. Several regularities that are species dependent are evident in the distribution of $^{239+240}Pu$ among different body tissues. Concentrations in liver always exceeded those in bone and concentrations were lowest in the muscle of all fish analyzed. A progressive discrimination against $^{239+240}Pu$ was observed at sucessive trophic levels at all atolls except Bikini and Enewetak, where it was difficult to conclude if any real difference exists between the average concentration factor for $^{239+240}Pu$ among all fish, which include bottom feeding and grazing herbivores, bottom feeding carnivores and pelagic carnivores from different atoll locations. The average concentration of $^{239+240}Pu$ in the muscle of surgeonfish from Bikini and Enewetak was not significantly different from the average concentrations determined in these fish at the other, lesser contaminated atolls. Concentrations among all 3rd, 4th, and 5th trophic level species are highest at Bikini where higher environmental concentrations are found. The reasons for the anomalously low concentrations in herbivores from Bikini and Enewetak are not known.

1. INTRODUCTION

One of the consequences resulting from the disposal of any radiological wastes into the sea is that a fraction of the radionuclides, derived from the wastes, may accumulate in a variety of marine organisms used as dietary items by man. Plutonium isotopes and other transuranics are among the longer lived radionuclides associated with different sources of nuclear wastes and it has been well documented that these radionuclides are readily concentrated by marine biota.

During the last decade much information has accrued on the concentrations and distributions of the transuranics in a variety of marine organisms from several environments contaminated by various sources of radioactive wastes. Many useful relationships have been developed from these studies but an examination of the literature reveals there are a sufficient number of unexplained differences in the available data to show that our experience in marine studies of the transuranics has still been too brief to model or to predict confidently concentrations or pathways of transuranic uptake by fish. Consider, for example, some results from several recent studies related specifically to the concentrations of plutonium accumulated by different marine vertebrates. The concentration factor (CF), a term assumed to be single valued by many, determined for plutonium in the flesh of different species of fish, abstracted from several reports [1,2,3,4,5,6,7], ranges in value from 0.04 to 1.3 x 10^3. It can be argued that different CF should be expected among different species with different feeding habits. However, even for the same species, the CF for plutonium varies significantly. Reported concentration factors for plutonium in the muscle of plaice, *Pleuronectes platessa*, range from 0.04 to 73 [3,4,8,9]. Similar differences are reported in fresh water where CF range from 6 to 235 for plutonium in smelt, *Osmerus mordax*, from four of the Great Lakes in the United States [10]. It would be difficult to select a single value among these results to predict fish concentrations from environmental data.

Studies also indicate that the relationship between the concentration of plutonium in marine species and the trophic level of the organism has not been completly resolved. Guary and Fraizier [4] show a decrease in concentration of plutonium as the trophic level of the organism increases, but data from Neyessi and Schell [6] indicate the opposite. They report [6] a CF of 1.5 x 10^3 for plutonium in the dark muscle of a skipjack tuna (4th or 5th trophic level [11]) from Enewetak Atoll and a CF of less than 20 in the muscle of surgeonfish (2nd trophic level [11]) from Bikini. Livingston and Bowen [12] conclude that the greatest influence of trophic levels on plutonium uptake by fish is related only to contact with bottom sediments. Differences have also been observed in the distribution of plutonium in fish tissues. Several studies show that the concentration of plutonium in muscle tissue is substantially lower than the concentrations in either bone or liver [1,3,9] but others [13,14,15] provide results which occasionally show an opposite trend in the enrichment pattern of plutonium by these tissues.

Some studies show that the concentration of plutonium in fish is related to concentrations in the environment but results from other research show that the concentrations of plutonium in tissues may be independent of the concentrations in the environment. Schell and Watters [14], for example, report finding concentrations of plutonium in the surgeonfish from Enewetak Atoll not significantly higher than concentrations measured in fish from their control station, Kwajalein Atoll. Samples of seawater and sediment from Kwajalein lagoon contained concentrations of plutonium several orders of magnitude lower than concentrations at Enewetak [5,16]. In one of four unrelated studies with fish having similar feeding habits, Bowen, et al., [13] reported a plutonium concentration of 0.08 + .04 pCi/kg in the flesh of menhaden collected from a global fallout contaminated environment in the North Atlantic. An average concentration of 0.10 pCi/kg was measured in the flesh of flatfish collected offshore from the Tokai fuel reprocessing plant in Japan [7]. Pentreath and Lovett [3] reported a concentration of .09 + .02 pCi/kg in the muscle of plaice collected during Nov. 1976 from the vicinity of a nuclear fuel reprocessing plant. Filtered seawater samples taken during the previous 3 months from the region (but not necessarily from the same water mass in which the plaice were sampled) averaged 2070 fCi/l. Guary, et al., [9] report a concentration of 0.11 pCi/kg in the flesh of plaice collected inshore near the La Haque fuel reprocessing plant where seawater concentrations were 13 fCi/l. The similarity in concentrations of plutonium detected in the muscle of these fish caught from four different environments, contaminated with very different levels of plutonium, suggests again the possibility that muscle tissue concentrations may be independent of the environmental concentrations.

These few examples cited serve to emphasize that our current understanding of the behavior of transuranics in biotic components of the marine environment is still inadequate in many areas and more information is needed before satisfactory models can be developed to relate environmental concentrations to the concentrations accumulated by marine vertebrates.

During the past several years we have collected a number of marine organisms, including species of fish, from atolls in the northern Marshall Islands for radionuclide analysis. The impetus for obtaining this data was to establish an updated data bank to assess any potential radiological consequences to man from ingestion of indigenous marine foods. In addition these measurements have provided data which may be useful to better understand the behavior of the transuranics and other radionuclides in the environment. In this report we discuss the portion of our data related to transuranic concentrations in marine vertebrates from the atolls of the northern Marshall Islands. We will attempt to emphasize regularities and differences that the results provide with respect to tissue distributions, trophic level relationships, feeding habits, environmental concentrations, and to other studies of plutonium in contrasted marine environments.

2. GENERAL INFORMATION

2.1 Background

Bikini and Enewetak Atolls were the locations of 22 and 43 nuclear tests, respectively, conducted by the United States between 1946 and 1958. Radioactive fission and activation products including the transuranium elements were generated during testing and deposited as close-in fallout on the atolls. Since 1972 we have been conducting studies at Enewetak and Bikini to better define the physical, chemical and biological transport mechanisms and fate of the transuranics and other long-lived radionuclides in the aquatic environments. Data are used to assess concentrations in different indigenous marine food products. Fish are a major component of the diet of Marshallese people throughout the Islands. The average daily consumption rate can vary from less than 200 g/day to over 600 g/day [17, 18]. Part of our study has therefore involved measuring concentrations of different radionuclides accumulated by fish and other marine organisms.

Other atolls in the northern Marshall Islands were contaminated to lesser and different degrees by intermediate-range fallout from one or more of the nuclear tests conducted at Bikini and Enewetak. In 1978 a radiological survey of 10 atolls and 2 separate islands in the northern Marshalls was conducted to document the amounts of man-made radioactivity remaining at these atolls and to assess radionuclide concentrations in different indigenous food products. A comprehensive survey of Bikini was also accomplished during this time. As part of the survey effort over 4000 fish were collected for analysis. The atolls and islands visited during the 1978 survey, in addition to Bikini, are shown in Figure 1 and include Utirik, Rongelap, Ailuk, Likiep, Wotho, Ujelang, Mejit, Bikar, Taka, Rongerik, Ailinginae and Jemo. The first 7 atolls are at present inhabited.

2.2 Collection methods

Throw nets were exclusively used to catch reef fish from the locations indicated by different letters and numbers in Figure 1. Large pelagic and benthic fish were collected on sport fishing gear, using feathered jigs, while trolling in the lagoons. The fish were returned to the research vessel, segregated by species, placed in plastic bags and frozen in freezer units. The samples were shipped frozen to Lawrence Livermore National Laboratory (LLNL) for storage and eventual processing.

2.3 Species collected, feeding habits and trophic level relationships

The principal species collected in 1978 were selected because they are commonly eaten by the Marshallese; relatively abundant at all atolls and at different locations within a atoll; have different feeding habits; and for some, represent species for which previous radiological data was available at Enewetak and Bikini for comparison. It was not always possible to obtain a sufficient number of the same species at every location we sampled. The reef fish collected at most atolls and for which data is provided in this report include:

Two species of mullet, Crenimugil crenilabis and Neomyxus chaptali. Mullet are herbivorous and detrital feeders. Considerable quantities of bottom sediment are ingested along with food. Adult mullet belong to the 2nd trophic level [11].

Surgeonfish, Acanthurus triostegus These fish are herbivorous browsers, feeding on algal fronds and filamentous algae which grow on reef rock or on the base of dead coral. This species is in the 2nd trophic level [11].

Goatfish, Mulloidichthys samoensis. Goatfish consume fossorial as well as surface benthic fauna including small clams, crustaceans, othjer invertebrates and small benthonic fish. The species belong to the 3rd trophic level [11].

The larger benthic, mid-water and surface carnivores occasionally collected from the lagoons included:

Groupers, Epinephelus sp., which are benthic carnivores feeding on small fish and invertebrates and are listed in the 3rd trophic level [11].

Jacks, Caranx melamphgus and Elegatis bipinnulatus, are fast swimming carnivores which feed on small fish and squid. Elegatis may occasionally eat swimming crustacea. Jacks are in the 4th marine trophic level [11].

Snappers, Aprion virescens and Lutjanus bohar. These fish are hovering mid-water to surface carnivores in the 4th marine trophic level [11].

Tunas and Mackerel, Euthynnus affnis, Thunnus albacare, Gymnosarda nuda and Grammatorcynus billineatus. These are large rapid swimming carnivores feeding on small fish and any prey of the proper size and represent species in the 5th trophic level.

2.4 Sample processing and analysis

The sorted species from each location were numerically counted and partially thawed. The total weight and standard length of each fish was usually determined. Each fish was opened to determine it's sex and then dissected into different parts. Each separated tissue and organ of the species from the same catch was pooled. Gills were separated from the 1978 collections but not analyzed. Our experience prior to 1978 showed the gills were often contaminated with sediment which contained significant quantities of radionuclides. The gills are not eaten and there could be little academic information gained from their analysis because of the probable contamination.

Samples were either sent to a contractor laboratory for analysis or processed and analyzed at LLNL. A complete description of the field program, sample processing, types and numbers of fish analyzed and analytical procedures are discussed in a report currently in preparation [19]. Plutonium was separated from ashed samples using a published technique [20] and finally electroplated onto stainless steel disks and measured by alpha spectrometry.

A number of duplicates, blanks and standard samples were intermingled with the regular samples analyzed here and at the contractor laboratory. All available quality control results for the fish samples demonstrated that the analytical performance was extremely good. A full discussion of all the quality control data for the 1978 survey results is also in preparation by others.

2.5 Environmental Concentrations

Water and sediment samples were also collected from regions of the lagoon reef at the atolls sampled in 1978. Average concentrations of $^{239+240}Pu$ in the lagoon water are given in Table I along with mean concentrations in surface water samples from the North Equatorial Pacific. Previously reported concentrations measured in lagoon water from Bikini and Enewetak are also shown. Table II lists surface sediment concentrations of $^{239+240}Pu$, ^{238}Pu and ^{241}Am along with previously reported results from Enewetak and Bikini.

All water samples collected from the lagoons of Enewetak and Bikini show $^{239+240}Pu$ concentrations greatly in excess of the average concentration in the surface water of the North Equatorial Pacific which is the original source for the lagoon water [21, 22]. These results demonstrate that $^{239+240}Pu$ is continuously mobilized to solution from the solid phases of the marine environments. Table I shows that average total concentrations of $^{239+240}Pu$ in the lagoon water from several other atolls in the northern Marshalls also exceeds the average concentrations in surface waters outside the lagoon. Mobilization of plutonium from sediments to water is evident in many lagoons of the northern Marshalls.

3. RESULTS AND DISCUSSION

3.1 General

Over 4200 fish have been processed since 1974 and more than 1100 samples of pooled fish parts were generated and analyzed for plutonium, americium and other radionuclides. Fewer determinations of Cm activities were made and these only in fish from Bikini and Enewetak. It was originally intended to present and discuss all available results for the different transuranics but as this report developed it became clear that the number of results and necessary discussions could not be adaquately condensed to a length suitable for these proceedings. It was therefore necessary to revise our original intention and eliminate all discussions related to transuranics and other radionuclides except plutonium. It was also necessary to limit the amount of plutonium data presented. In Tables III and IV a representative fraction (less than 25%) of the concentrations of $^{239+240}Pu$ in fish from Bikini, Enewetak and other atolls are shown. These were selected to illustrate certain relationships discussed in the text. Other tables in this report will summarize data appearing in these tables along with our unpublished results. We emphasis this because it may not be obvious how some of the average concentrations or concentration quotients have been determined from only the results shown.

Some of the material eliminated from the discussion deserves mention in a condensed form. Throughout the Marshall Islands, transuranic dose rates to man from consumption of fish muscle tissue are less than the 3 mrad/yr guideline recommended by the Environmental Protection Agency of the United States [23]. If other parts of the fish are eaten, especially at Bikini and Enewetak, dose rates could exceed this guideline. Concentrations of plutonium in most fish parts from any one location collected in different years from either Bikini or Enewetak have comparable concentrations (see samples 5286, 9115 and 7247, 2851 in Table III for examples). These and other results show that reef species have restricted feeding territories within a lagoon. Curium-242,243,244 have been detected in some fish tissues from Enewetak and Bikini. The concentrations vary with the geographical region of the atoll from where the fish were caught. Concentrations of $^{243,244}Cm$ are a few percent of the $^{239+240}Pu$ concentrations and ^{242}Cm is less than 1% of the $^{239+240}Pu$ levels in the entire fish. The detection of ^{242}Cm ($t_{1/2}$=163d) in environmental samples, 20 years after the end of testing, indicates the presence of a parent radionuclide, ^{242m}Am. The food chain behavior of plutonium does not parallel that of ^{137}Cs. ^{137}Cs concentrations are lowest in bottom feeding fish and increase in concentration with trophic level except that surgeonfish have concentrations higher than one would predict from its position in the trophic scale. The reverse trophic relationship is found for $^{239+240}Pu$. If the body burdens of transuranics are accumulated by the fish through the gut, the results indicate that americium is transferred relatively less efficiently across the gut than is plutonium. The concentration ratio of ^{241}Am: $^{239+240}Pu$ in muscle, bone, skin or liver was always either equivalent to or less than the ratio in the material ingested. Pentreath and Lovett [3] however, found exactly the opposite to be the case for plaice collected from vicinities near the Windscale fuel reprocessing facility. Ratios computed from their concentration data or concentration factors show, in 9 different monthly collections of fish, that the activity ratio is lowest in the ingested material relative to the ratio in other parts of the fish. Clearly more information is required before the biokinetic behavior of the transuranics in marine vertebrates is understood. Some of these subjects will be discussed in future reports.

3.2 Tissue and Organ Concentration of $^{239+240}Pu$

There are several regularities in the data related to the distribution of $^{239+240}Pu$ among different organs of all fish. These few generalized observations apply to all species from every trophic level collected from every atoll contaminated with different levels of activity.

a. The concentration of plutonium in the pooled muscle tissue was less than the concentration measured in the bone and liver of the fish.

b. Concentrations in liver were always higher than the levels detected in bone.

Some regularities were evident which appear to be independent of geography or level of contamination but dependent on the species. Table V shows several average values of the plutonium concentration ratio among tissues of the different fish.

c. The muscle to bone ratio is smallest in surgeonfish and increases in value with trophic level although more variability was encountered in generating a mean value for this quotient in goatfish and other carnivores. It was surprizing to find a significant difference in this ratio for the two species of mullet. Not only are differences noted when comparing species but some differences are encountered among species from the same family.

d. The skin to bone ratio was relatively constant among all fish collected in 1978 but the quotient varied significantly among the larger carnivores and among reef fish collected at Enewetak and Bikini during 1976 and 1977. We attribute this to different sampling techniques. In 1976 and 1977 the reef fish were extracted from throw nets and immediately (within 15 minutes) returned to the research vessel, rinsed clean with fresh water, segregated by species, placed in plastic bags and frozen. In 1978 small boats were in demand for other operations during the survey and could not stand by to retrieve fish samples. To keep the fish as fresh as possible it was necessary to transfer the live catch to buckets containing seawater. The water was continuously replaced. When a boat arrived, the fish were segregated and transferred to ice chest. All subsequent procedures used to clean, dissect and analyze all fish collections were identical. By leaving the live fish in buckets of seawater they were able to purge themselves of some plutonium adhering to the scales and skin. It was also noted that a number of scales were lost from the fish during confinment in the buckets. A loss of scales or the associated mucous would also account for a lower skin-scale to bone concentration ratio. Scales were separated from the skin of mullet collected from Enewetak in March 1978 and contained 5.7 pCi/kg of $^{239+240}Pu$ while the skin concentration was 1.1 pCi/kg. The larger fish, with the variable concentration ratios in Table V, were caught while trolling in the lagoon from the small boats. After capture the fish were immediately transferred to ice chests, duplicating, essentially, the collection procedure used prior to 1978. Different concentrations of plutonium may be encountered among fish tissues exposed to the environment as a result of collection and handling techniques.

e. Liver to bone concentration ratios were variable but concentrations in all liver samples were higher than the respective concentration in bone. In laboratory experiments with plaice, Pentreath [24] found the quotient of plutonium activities in liver to bone ranged from values greater than 30 to less than 10 and decreased as a function of the fishes growth rate. We attempted to correlate the liver to bone concentration ratio with the mean standard length of the pooled mullet samples but found no satisfactory relationship between activity ratio and size. In fact, there was a larger fraction of higher activity ratios associated with larger sized fish which is the reverse of the observation made with plaice [24].

f. The average quotient of the viscera to stomach content concentrations were unique to the species. In both species of mullet the average concentration of $^{239+240}Pu$ in the stomach contents exceeded the concentration in the viscera while the goatfish viscera concentrations always exceeded the level in the stomach contents.

g. The average concentration of $^{239+240}Pu$ in the eyes of surgeonfish was 20 times the concentration in muscle tissue and always lower than the bone concentrations.

Some regularities were found only among fish from specific atolls.

h. At Bikini and Enewetak, the gut content in the two species of mullet contained the highest concentration of $^{239+240}Pu$ detected in fish parts, as would have been anticipated for a bottom-feeding fish.

i. Bone to stomach content concentration ratios in fish from different atolls are also shown in Table V. There is a large difference between the ratio in fish from Enewetak and Bikini compared to the ratio in fish from other atolls. Concentrations of plutonium in bone and other internal organs are not proportional to gut content concentrations in a species from different atolls but some regularity in the ratio is evident in the species sampled at the same atoll.

A number on non-predictable irregularities were also found.

j. Using observation h, we would have predicted that the stomach content concentrations would be highest among other parts of mullet from other northern Marshall Atolls. Some mullet were found to contain concentrations in the liver which exceed the stomach content concentration (see samples 6358c and 6712c, Table IV as examples).

k. The concentrations in the intestinal contents of surgeonfish were inconsistent with respect to the concentration in the viscera. Unlike the consistent ratios found among the mullet and goatfish, concentrations in the viscera were found to be either higher or lower than concentrations in the intestinal contents.

l. Concentrations of $^{239+240}Pu$ in the reproductive organs of fish from Bikini and Enewetak were variable and unpredictable. There was no correlation between the concentrations in ovaries and testes of fish caught at the same time from the same location.

Although there are a number of regularities in the distribution of $^{239+240}Pu$ among fish parts which have not been previously reported, our data also provided a number of unpredictable irregularities which are, at present, difficult to explain.

3.3 Concentration factors and trophic level concentrations of $^{239+240}Pu$

The average amount of $^{239+240}Pu$ mobilized to lagoon water from sedimentary components at Enewetak and Bikini has been relatively constant for at least the last 16 years [21,22,25]. Biological species used as chronometers [25], or serially sampled from the same locations, show little variation in concentration with respect to time. These results indicate that the lagoons have approached, if not attained, a chemical steady state condition with respect to the partitioning of plutonium among the environmental components in the lagoon.

Concentration factors for plutonium in the muscle of mullet and surgeonfish were computed for collections made at Enewetak in 1976 using the mean concentration measured in filtered water collected near the islands where the fish were caught. Muscle and water concentrations and the computed concentration factors are given in Table VI. Concentrations of $^{239+240}Pu$ in water varied within the lagoon and this variation was reflected in the concentrations in fish. Concentration factors based on total plutonium in the water (soluble and particulate), would be less than the values shown. It was encouraging to find so little variation in the CF for a species over the entire lagoon considering the reef water in any region can vary significantly even over daily tidal cycles. The average CF for $^{239+240}Pu$ in the muscle of mullet was only slightly higher than the mean value in surgeonfish. A useful relationship was established at Enewetak to estimate intra-atoll concentrations but it remained to be established whether the CF was valid for the same species at other atolls. Table VI shows that a similar mean concentration factor was found for mullet from Bikini in 1977. In 1978, the mean values were again verified for these fish from both Enewetak and Bikini. Our mean CF in surgeonfish also agreed with the results of Nevessi and Schell [6] who reported a CF for $^{239+240}Pu$ of less than 20 in the muscle of surgeonfish collected at Bikini in 1972. After essentially duplicating the mean value of the CF for $^{239+240}Pu$ in muscle of mullet and surgeonfish at two atolls, over a 3 year period, a greater reliance was placed on its general usefulness to predict concentrations from environmental data. Good agreement was, in fact, found between measured and predicted concentrations in two fish from Kwajalein Atoll and in fish from other global locations [5]. It was therefore anticipated that similar values would be determined for $^{239+240}Pu$ in muscle of fish from the other atolls sampled in 1978. Many of the atolls were contaminated by the same sources and have had a similar time to reach steady state conditions.

A summary of mean concentrations for $^{239+240}Pu$ in fish muscle and concentration factors from the 1978 collections are shown in Table VII and ranked relative to the position of the species in the trophic level scale. At some atolls, different concentrations were evident in a species from different locations but the variation encountered over an atoll was much less than we found at Enewetak or Bikini. A concentration factor was determined from the water concentration at the site sampled or, when the concentration at a specific location was unavailable, from the average lagoon concentrations given in Table I. Mean lagoon water concentrations were used to compute CF for the larger predators. All values for the species in a lagoon were averaged. Inspection of this data reveals several interesting patterns and a number of unanticipated results.

a. There is good agreement among the values for the CF in the muscle of 4th and 5th trophic level fish from all atolls where average concentrations in the flesh range from less than 0.003 to 0.23 pCi/kg. An average value of 10 or less is in agreement with reported CF in other pelagic predators [1,4].

b. Except at Enewetak and Bikini, there is a decrease in the CF and concentration of $^{239+240}Pu$ in fish between the 2nd and higher trophic levels. This trend is in agreement with trophic level relationships previously described for marine and fresh water [4,26].

c. The mean CF for $^{239+240}Pu$ in 3rd trophic level, bottom feeding carnivores, was not substantially different from the value for mid-water feeding carnivores. On the other hand, except at Likiep, the CF for the 2nd trophic level, bottom feeding mullet, was everywhere higher than the value for surgeonfish.

d. In contrast to finding a dependence of concentration with trophic level, it is difficult to conclude that any real differences exist at Enewetak and Bikini between the average CF for $^{239+240}Pu$ in all fish which include bottom feeding and grazing herbivores, bottom feeding carnivores and pelagic carnivores from different lagoon locations.

e. The mean CF for $^{239+240}Pu$ in muscle of mullet and surgeonfish were significantly less at Bikini and Enewetak compared to the mean value for the species at other atolls. Based on the results from Bikini and Enewetak, predicted concentrations in these fish from other atolls would have differed from the measured concentrations by factors greater than 10. It was anticipated that with this body of data more regularities would be obvious than we find in the values of CF for a single species from the different atolls.

f. Table VI shows that concentrations of $^{239+240}Pu$ in the muscle of surgeonfish from locations within Enewetak lagoon range from 0.43 to 0.008 pCi/kg. Highest concentrations were twice detected in fish from island E-2, located in the northeast region of the atoll near 2 large nuclear craters. If these highest concentrations are eliminated from the atoll average value given in Table VII, the mean concentration in these fish from the remainder of the atoll is 0.065 pCi/kg. A concentration equivalent to or larger than this average was measured in 17 of the 28 surgeonfish collections from the lesser contaminated northern Marshall Islands. At Enewetak the lowest concentrations of $^{239+240}Pu$ in muscle were measured in surgeonfish and mullet collected from island E-37. The water concentrations in this region of Enewetak lagoon have been consistently higher than concentrations in water from all other atolls except Bikini. However the muscle concentrations of $^{239+240}Pu$ were higher than the concentrations in the respective species from E-37 in 25 of the 28 collections of surgeonfish and 19 of the 22 collections of mullet from the lesser contaminated atolls. It was unanticipated to find the average concentration for $^{239+240}Pu$ in muscle of mullet from Rongelap Atoll larger than the average concentrations from all other atolls, including the more contaminated atolls of Bikini and Enewetak. The generalization that can be made from these comparisons is that concentrations of $^{239+240}Pu$ in the muscle and other tissues of some species are not necessarily related to the concentrations in the environment from which they are obtained. It may be argued that some of these paradoxical findings result from uncertainties associated with measuring low concentrations of $^{239+240}Pu$ in muscle. But recall that consistent relationships were found between the concentrations in muscle and bone of a species from all atolls. Concentrations in bone are orders of magnitude larger than in muscle. Tables III and IV show that many concentrations in bone of surgeonfish and mullet from Bikini and Enewetak are low compared to, for example, bone concentrations in fish from Rongelap in spite of the fact that concentrations in water, sediment and material ingested by fish are orders of magnitude higher at Bikini and Enewetak. The consistency of measured concentrations rules out analytical or sampling errors to explain these findings. No relationship between the size (assumed to be related to age) of a species and concentration could be developed from the results nor were there correlations with season of the year sampled.

If the concentrations in herbivores from Bikini and Enewetak are anomalously low, then there must be a significant difference in the bioavailability of $^{239+240}Pu$ to these fish from either the water or ingested material everywhere in the lagoons. Why this should be and only at Bikini and Enewetak is difficult to understand.

Absorption from the water does not appear to be a major pathway for uptake of $^{239+240}Pu$ by mullet or surgeonfish but it may be for other fish. We have a sufficient amount of data from Enewetak and Bikini to show that from 75 to 94% of the soluble $^{239+240}Pu$ in the lagoon water is in the +5 or +6 state. The remainder is in either the +3 or +4 state. All plutonium associated with particulate material in the water column is in the reduced states. Concentrations of either oxidized or reduced pair in the water from the majority of locations sampled at Bikini and Enewetak exceed the total concentration of soluble plutonium at the other atolls. If there are differences in the absorptive capacity for $^{239+240}Pu$ in different oxidation states, concentrations in mullet and surgeonfish should always be highest everywhere at Enewetak and Bikini.

Mullet and surgeonfish from Enewetak and Bikini pass material with considerably higher concentrations of $^{239+240}Pu$ through their gut than do these fish from other atolls but fail to reflect these higher concentrations in muscle, bone and other tissues. Observations made during dissections showed the contents from all mullet consisted of sediment and unidentified organic matter. The wet/dry weight ratio for the contents from mullet was a constant at all atolls and averaged 1.6 ± 0.2. A constant ratio of 6.7 was found for the contents from surgeonfish. The different values attest to the different feeding habits of the two species and the observations indicate the species from all atolls are probably feeding on similar material. This contention is supported by data in Table VIII which shows reasonable agreement between the concentrations of $^{239+240}Pu$ associated with the material ingested by mullet and the near shore sediments. It is possible that only a much smaller fraction of the total $^{239+240}Pu$ associated with the material ingested from anywhere in the lagoon at Bikini and Enewetak is exchangeable and available for absorption through the gut compared to the amounts mobilized in the guts of fish elsewhere.

At Enewetak and Bikini the average quantity of $^{239+240}Pu$ mobilized to the water column is proportional to the average concentration in the lagoon surface sediments [22]. A number of laboratory and in situ experiments have been conducted with contaminated sediments to arrive at a value for the distribution coefficient (K_d) for plutonium. The range in K_d values determined for the different lagoon sediments from Bikini and Enewetak was between 0.5×10^5 and 3.6×10^5 with an average K_d value for $^{239+240}Pu$ of 2.3×10^5. The average concentration of $^{239+240}Pu$ in the lagoon water is related to the K_d value and the average sediment concentration. If this relationship holds at other atolls, average water and sediment concentrations can be used to estimate a value for the distribution coefficient. This value can be useful to assess if more or less plutonium is capable of dissociating from the lagoon sediments. Average K_d's are computed at several atolls using average concentrations in sediment, material ingested by mullet, and average water concentrations. Results are shown in Table VIII. The range encountered in values at Enewetak is similar to the range in the K_d values at other atolls. These comparisons indicate that similar fractions of plutonium are mobilized to solution from the sediments at all atolls. It may be assumed for any atoll that a proportional fraction of plutonium is desorbed from the material as it passes through the gut of the fish. The differences in muscle concentrations cannot be accounted for by assuming any less a fraction of the plutonium in the material ingested by fish at Bikini and Enewetak is available for absorption across the gut.

4. CONCLUSIONS

In a previous publication [5] we reported that the CF for $^{239+240}Pu$ appeared to be independent of species, trophic level and location based on limited data obtained at two Pacific atolls. In contrast, our present results show that the CF for $^{239+240}Pu$ in tissues of some species shows a definite geographical dependence, appears to be related to the species, and, at some atolls, may or may not be trophic level dependent. Our current understanding of the factors which influence uptake of $^{239+240}Pu$ by some species at at atoll is not adaquate to explain the present results. This and other studies show that it may be becoming a rule, rather than an exception, to find concentrations of $^{239+240}Pu$ in the tissues of some fish unrelated to the environmental concentrations. Therefore limited use should be made with a CF for $^{239+240}Pu$ in situations which require extrapolating values determined in one environment to estimate impacts on the same or different fish in other environments.

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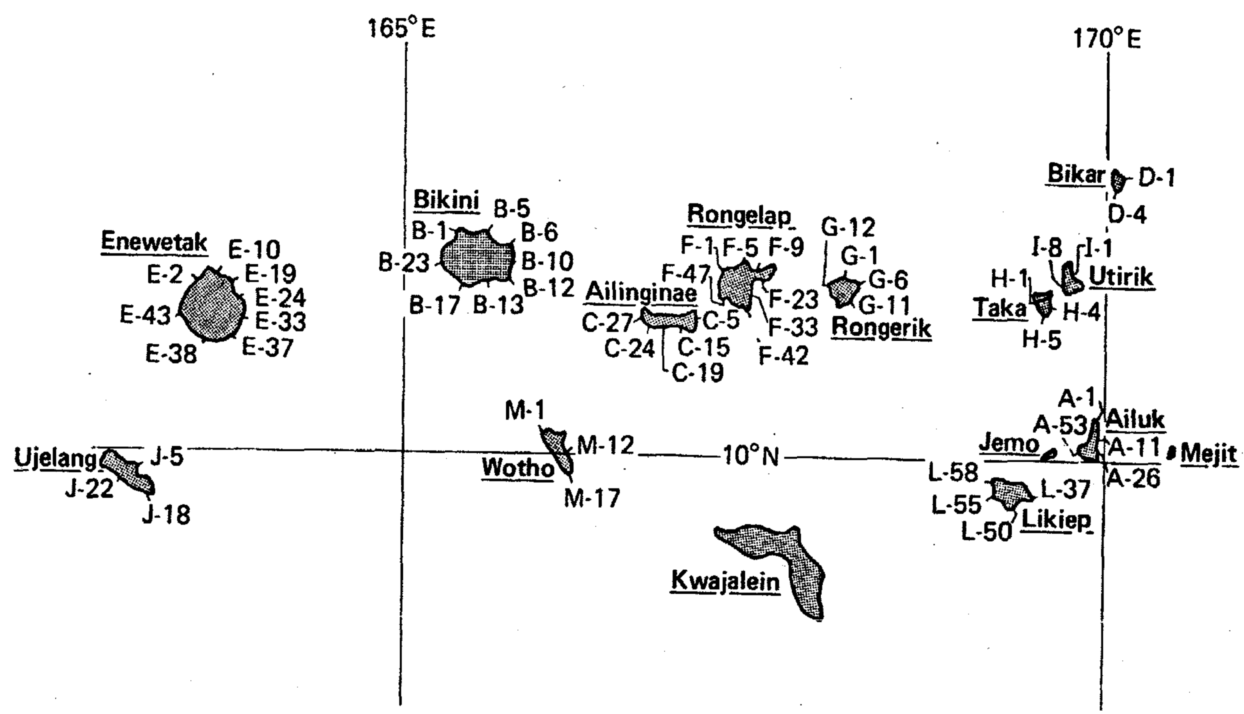

FIG. 1. Atolls and locations sampled in the northern Marshall Islands.

TABLE I

Mean and range of $^{239+240}Pu$ concentrations in lagoon seawater from atolls in the northern Marshall Islands.

Atoll	Number of samples	Year collected	$^{239+240}Pu$, pCi/m^3 Soluble[a]	Particulate[a]	Mean Total
Ailinginae	5	1978	0.48(.25-.99)	0.18(.06-.31)	0.66
Ailuk	4	1978	0.57(.18-.95)	0.04(.01-.07)	0.61
Bikar	2	1978	0.38(.34-.41)	0.80(.05-1.5)	1.18
Jemo	1	1978	0.64	0.09	0.73
Likiep	4	1978	0.58(.24-1.0)	0.24(.02-.66)	0.82
Mejit	1	1978	0.49	0.26	0.75
Rongelap	6	1978	1.31(1.1-1.7)	1.33(.11-2.9)	2.64
Rongerik	4	1978	1.20(.20-3.4)	0.15(.02-.31)	1.35
Ujelang	4	1978	0.18(.13-.33)	0.13(.03-.34)	0.31
Utirik	3	1978	0.26(.16-.35)	0.14	0.40
Wotho	4	1978	0.42(.14-.56)	0.03(.01-.06)	0.45
Enewetak[b]	48	1974	24(1.4-65)	19(1-125)	43
Enewetak	19	1976	17(2-31)	13(2-67)	30
Enewetak	5	1978	17(3-46)	19(1-40)	36
Bikini[b]	17	1972	40(4-79)	36(0.1-460)	76
Bikini	26	1977	49(13-104)		
Bikini	8	1978	29(7-50)	40(3-98)	69
North Equatorial Pacific Surface Water[b]	14	72-78		-	0.38

a - Values in parenthesis represent the range in concentrations.
b - From refs. 21 and 22.

TABLE II

Mean and range of $^{239+240}Pu$, ^{238}Pu, and ^{241}Am concentrations in lagoon surface sediments (4 cm) from atolls in the northern Marshall Islands.

Atoll	pCi/kg dry weight[a] $^{239+240}Pu$	^{238}Pu	^{241}Am
Ailinginae	60(26-85)	0.7(<.4-1.0)	43(19-55)
Ailuk	9(5-13)	0.4(<.4-.6)	8(5-10)
Jemo	13	1.0	12(11-14)
Likiep	7(3-11)	0.4	7(4-8)
Rongelap	280(115-525)	1.0(<.2-2.0)	196(98-350)
Rongerik	86(29-167)	0.8(<.2-1.8)	34(16-70)
Taka	13(7-18)	0.7(<.2-1.2)	11
Ujelang	20(16-30)	0.3(<.2-.6)	14(8-22)
Utirik	16(10-25)	0.4(<.1-0.7)	13(5-24)
Wotho	10(6-13)	0.8(<.1-1.5)	8(3-12)
Enewetak[b]	5200	732	2084
Bikini[b]	9500	491	7360

a - Values in parenthesis represent the range in concentrations.
b - From refs. 21 and 22.

TABLE III

Concentration of $^{239+240}Pu$ in of reef fish from Bikini and Enewetak Atolls.

Sample I.D.	B - Bikini E - Enewetak	Month/yr collected	Tissue and organs analyzed - $^{239+240}Pu$-pCi/kg wet weight[a]								
			Muscle	Bone	Stomach contents	Viscera	Scales & skin	Liver	Gill	Ovary	Testes
7257-S[b]	B-5	11/78	.12(29)				10.0(10)	73(7)			
7269-S	B-10	11/78	.11(20)		150(5)			27(4)			
5286-S	E-2	4/76	.43(5)	25.4(8)	d	950(6)	14.3(2)	c	20(7)	167(3)	55(5)
9115-S	E-2	11/78	.40(9)	25.2(6)	510(2)		16.7(2)				
5270-S	E-19	4/76	087(10)	8.1(2)	d	280(10)	6.3(18)	c	1.2(30)	23.4(3)	42(10)
5294-S	E-24	4/76	.078(13)	6.9(3)	d	380(10)	3.6(5)	c	6.4(6)	5.7(46)	2.8(23)
5239-S	E-37	4/76	.008(25)	0.2(14)	d	8.7(3)	0.2(11)	c	0.9(10)	0.5(17)	
5247-S	E-38	4/76	.037(16)	1.8(9)	d	69(3)	1.0(12)	c	7.9(6)	2.4(10)	2.23(11)
9133-C	B-1	11/78	.64(5)	60(3)	5200(20)		18.7(2)				
2880-C	B-2	1/77	.54(4)	23.9(1)	17200(7)	4300(6)	56(26)	c	45(24)	11.8(5)	134(5)
7245-C	B-5	11/78	.28(7)	24.8(6)	6000(3)	1700(20)	8.6(6)	527(2)			
2860-C	B-12	1/77	.13(7)	4.0(3)	600(30)	220(2)	41(10)	c	7(25)	3.2(6)	
2851-C	B-13	1/77	.089(7)	4.1(3)	800(4)	136(3)	8.5(19)	c	6(25)	3.6(22)	
7212-C	B-13	11/78	.083(7)	5.3(4)	810(3)	300(2)	2.3(6)	202(2)			
2610-C	E-2	4/76	.38(5)	4.9(2)	14200(4)	1700(5)	5.9(6)	c	15.0(5)	24.7(1)	
5302-C	E-10	3/78	.087(8)	7.45(10)	5800(2)	1160(1)	6.8(3)	64(2)	18.8(2)		
2625-C	E-37	4/76	.019(37)	0.34(15)	160(7)	53(7)	0.13(23)	c	0.8(10)	.07(21)	
2594-C	E-43	4/76	.18(17)	5.6(10)	650(2)	220(13)	2.5(3)	c	9.2(3)	6.8(8)	5.1(8)
2872-N	B-17	1/77	.87(2)	9.7(5)	1270(7)	1200(2)	7.4(13)	c	33(6)	25.8(6)	10.7(15)
7305-N	B-23	11/78	1.04(3)	35(4)	3000(20)	1750(2)	12.2(5)	1200(2)			
9103-N	E-2	11/78	1.47(3)	24.6(3)	9000(20)		9.7(6)	419(2)			
2641-N	E-19	4/76	1.60(5)	13.0(3)	80000(5)	49000(10)	7.7(4)	c	40(3)	632(4)	93(3)
2618-N	E-24	4/76	.40(5)	3.3(9)	6000(4)	377(6)	1.1(10)	c	160(8)	4.9(5)	
7206-G	B-13	11/78		1.6(7)	1.7(40)	5.9(4)	1.02(3)	39.3(3)			
7281-G	B-17	11/78	.073(23)		65(15)		3.4(8)				
9121-G	B-1	11/78	.18(24)	7.0(8)		73.6(3)	4.7(9)	175(5)			

a - Value in parenthesis is the percent standard deviation of the counting error.
b - Common Names: S - Surgeonfish; C - Mullet (Crenimugil); N - Mullet (Neomyxus); G - Goatfish.
c - Liver included with viscera sample.
d - Contents included with viscera sample.

TABLE IV

Concentrations of $^{239+240}Pu$ in reef fish collected between September-November 1978 from the Northern Marshall Islands.

Sample I.D.	Atoll	Island sampled	No. of fish[b] pooled	Average length (mm)	Average whole-body wt (gm)	Tissues and Organs Analyzed-$^{239+240}Pu$-pCi/kg wet weight[a]					
						Muscle	Bone	Stomach contents	Viscera	Scales & skin	Liver
6381-S[c]	Likiep	L-55	10(6)	113	82	.07(60)	4.3(13)	--	6.2(8)	2.4(13)	--
6437-S	Taka	H-5	25(24)	93	39	.12(8)	12.1(8)	--	10.8(6)	7.8(8)	--
6485-S	Ailinginae	C-27	73(51)	100	53	.11(21)	17.0(7)	5.3(12)	9.8(7)	7.9(6)	--
6790-S	Rongerik	G-1	64(61)	113	72	.15(13)	18.2(4)	8.6(7)	13.3(4)	14.7(5)	--
6971-S	Jemo	S-1	69(33)	125	98	.04(20)	7.6(3)	2.9(3)	3.7(2)	4.5(2)	29(3)
7029-S	Rongelap	F-1	51(50)	86	30	.69(11)	117(4)	--	10.3(4)	89(3)	--
7035-S	Rongelap	F-42	45(33)	112	73	.06(23)	10.8(6)	12.(10)	8.9(3)	8.1(6)	--
6358-C	Likiep	L-55	7(5)	243	406	.04(19)	1.9(12)	9.0(7)	3.5(2)	.34(18)	15.6(4)
6501-C	Ailinginae	C-5	5(3)	257	410	.14(7)	13.9(6)	237(2)	134(2)	4(4)	117(7)
6712-C	Ailuk	A-1	7(5)	186	164	.020(27)	3.3(8)	11(7)	--	1(10)	24.6(7)
7080-C	Rongelap	F-23	12(9)	261	472	.54(7)	43(4)	427(4)	217(4)	16.1(6)	--
7074-C	Rongelap	F-47	12(4)	298	664	.34(9)	16.2(5)	364(5)	153(3)	5.8(6)	176(2)
6449-N	Taka	H-1	34(33)	207	182	.19(9)	4.5(9)	18(5)	8.6(4)	1.0(8)	25(4)
6461-N	Taka	H-4	20(11)	204	161	.13(17)	2.8(9)	18(7)	12.5(5)	.91(10)	--
6574-N	Wotho	M-1	55(33)	184	130	.13(12)	3.1(6)	22(3)	3.5(3)	.84(8)	11.0(3)
6676-N	Bikar	D-1	7(1)	198	197	.016(30)	1.1(15)	76(3)	8.3(7)	.35(12)	9.4(7)
6531-G	Ailinginae	C-5	28(2)	199	163	.016(25)	.035(34)	--	5.5(8)	<.02	--
6730-G	Ailuk	A-53	23(17)	206	189	<.002	<.1	--	.3(32)	<.04	--
6350-G	Ujelang	J-5	26(10)	198	169	<.003	.13(50)	--	2.1(19)	<.03	--
6927-G	Utirik	I-1	76(73)	164	85	<.001	.05(50)	1(35)	2.8(2)	.03(60)	.6(26)
6467-H	Taka	--	2(0)	542	2618	<.004	.11(50)	--	.08(50)	.03	--
6945-E	Utirik	--	1(0)	378	853	<.01	<.01	--	--	.11(60)	--
6658-U	Bikar	--	2(1)	421	1440	.009(70)	<.02	--	<.03	<.07	--
7116-R	Rongelap	--	2(1)	538	1982	<.004	.10(80)	1(21)	.09(50)	<.07	--
7110-M	Rongelap	--	2(1)	518	1407	.009(43)	.03(50)	--	.13(40)	<.07	--
7149-B	Rongelap	--	1(1)	575	3883	<.005	.17(60)	0.6(57)	.13(45)	<.03	--
6832-T	Rongerik	--	2(0)	750	5773	.005(40)	<.02	.05	<.07	<.5	.19(36)

a - Value in parenthesis is the percent standard deviation of the counting error.
b - Number of male fish in parenthesis.
c - Common names: S - Surgeonfish; C - Crenimugil sp.; N - Neomyxus sp.; G - goatfish H - Snapper; E - Grouper; U - Jack (Ulua); R - Jack (rainbow runner); U - Mackerel; B - Bonito; T - Tuna.

TABLE V

Mean ratio of $^{239+240}Pu$ activity in fish tissues from all atolls and bone to stomach content concentration ratio at ~~specific atolls~~.[a]

Common name	$^{239+240}Pu$ Activity Ratios[a]			
	Muscle/bone	Skin/bone[b]	Liver/bone	Viscera/ stomach contents
Surgeonfish	.012±.008(36)	.56±.12(36)	5±2(2)	1.2±0.4(8)
Mullet (Crenimugil)	.020±.010(24)	.35±.07(18)	13±10(8)	.46±.14(9)
Mullet (Neomyxus)	.065±.043(17)	.32±.13(12)	14±11(8)	.49±.10(7)
Goatfish	0.13±.11(12)	.55±.19(7)	20±7(3)	3.2±0.5(4)
Large carnivores	0.12±.08(10)	variable (.2-4.6)	c	c

Bone/Stomach Contents

	Surgeonfish	Mullet
Bikini - Enewetak	0.07±0.04(6)	0.007±0.006(20)
All other atolls	1.8±0.9(7)	0.18±0.11(16)

a - Number of samples averaged are in parenthesis.
b - 1978 samples only.
c - Insufficient results presently available.

TABLE VI

Concentration factor (CF) for $^{239+240}Pu$ in fish muscle of Mullet and Surgeonfish from Bikini and Enewetak Atolls.

Year	Island	Average reef[a] water concentration pCi/kg wet	Mullet muscle concentration pCi/kg wet	CF	Surgeonfish muscle concentration pCi/kg wet	CF
1976	E-2	.037±.019(7)	0.38	10.3	0.43	12.0
	E-10	.033±.009(2)	0.41	12.0		
	E-19	.17 (1) ocean reef	1.60	9.2		
	E-19	.013 (1) lagoon reef	-	-	0.087	6.7
	E-24	.080±.030(12)	0.40	5.0	0.078	1.0
	E-33	.009 (1)	-	-	0.035	3.8
	E-37	.006±.003(5)	0.019	3.2	0.008	1.3
	E-38	.013±.006(5)	-	-	0.037	2.8
	E-43	.028±.008(5)	0.18	6.8	-	-
		yearly mean		8±3		5±4
1977	B-1	.036±.004(2)	0.38	10.6		
	B-2	.069 (1)	0.54	7.8		
	B-7	.027 (1)	0.13	4.8		
	B-10	.047±.006(2)	0.78	16.6		
	B-13	.009 (1)	0.089	9.9		
	B-17	.051(1)	0.87	17.1		
		yearly mean		11±5		
1978	B-1	.046 (1)	0.64	13.9		
	B-5	.027 (1)	0.28	10.4	0.12	4.4
	B-10	.008 (1)	-	-	0.11	13.7
	B-13	.042 (1)	0.083	2.0	0.039	0.9
	B-17	.030 (1)	0.28	9.3		
	E-2	.080 (1)	1.47	18.4	0.43	5.4
	E-24	.046 (1)	0.15	3.3		
		yearly mean		10±6		6±5
		overall average		9±5		5±4

a - Number of water samples averaged for mean concentration shown in parenthesis.

TABLE VII

Mean concentrations and concentration factors (CF)[a] for $^{239+240}Pu$ in muscle tissue of fish in different trophic levels[b] from different atolls.

Atoll	Primary consumers 2nd trophic level: Mullet	Primary consumers 2nd trophic level: Surgeonfish	Primary consumers 2nd trophic level: All	Secondary consumers 3rd trophic level (Goatfish-Groupers)	Tertiary consumers 4th trophic level (Jacks-Snappers)	Quaternary consumers 5th Trophic level (Mackerel-Tuna-Shark)
Ailinginae conc. (pCi/kg wet)	.20±.07(3)	.12±.05(4)	.15±.07	<.015(3)	<.008(2)	<.01(3)
CF	460±180	225±53	328±168	< 31	< 17	< 17
Ailuk conc. (pCi/kg wet)	.06±.04(2)	.05±.03(2)	.06±.03	<.006(2)		
CF	106±68	87±50	99±55	< 11		
Likiep conc. (pCi/kg wet)	.04±.01(3)	.06±.03(3)	.05±.02	.005±.003(2)		
CF	119±75	188±86	154±82	9±5		
Rongelap conc. (pCi/kg wet)	.72±.45(4)	.32±.21(7)	.35±.20	.034±.029(5)	.005±.002(2)	.008±.003(3)
CF	550±250	256±123	345±238	24±21	4±2	6±3
Rongerik conc. (pCi/kg wet)	.18±.15(2)	.09±.06(4)	.11±.09	.015±.010	.014±.008(2)	.005±.003(2)
CF	285±50	73±46	144±117	21±12	12	4±2
Taka conc. (pCi/kg wet)	.15±.03(3)	.08±.05(2)	.13±.05	-	.005±.002(2)	
Wotho conc. (pCi/kg wet)	.10±.05(3)	.04±.01(1)	.08±.05	.006±.005(2)	<.003(2)	
CF	228±140	77	190±140	14±12	< 6	
Bikini conc. (pCi/kg wet)	.45±.32(12)	.09±.03(3)	.38±.32	.13±.07(2)	.11±.01(1)	.23±.20(5)[d]
CF[c]	10±5	6±5	9±5	5±3	3	8±5
Enewetak conc. (pCi/kg wet)	.57±.61(8)	.15±.16(8)	.44±.55			
CF[c]	9±5	5±4	7±5			
Remaining atolls conc. (pCi/kg wet)	-	-	.04±.03(7)	<.004(5)	<.007(5)	
CF	-	-	102±75	10	< 18	
Mean CF - all atolls	-	-	194±101[e]	15±9	10±7	9±6

a - Values computed relative to filtrate water concentration in the vicinity where fish were caught; if unique concentration was not available mean lagoon concentrations from Table I were used.
b - Numbers in parenthesis are the number of samples averaged.
c - For mullet and surgeonfish - mean values from Table VI.
d - Includes results for shark collected in 1972.
e - Excluding Bikini and Enewetak.

TABLE VIII

Comparison of concentrations for $^{239+240}Pu$ in material ingested by mullet and in surface sediments from different atolls.

Atoll	Island I.D.	Ingested material $^{239+240}Pu$ mean pCi/gm dry	concentration pCi/gm dry range	mean	Average water concentration fCi/l	Kd x 10^{-5} [a] Ingested material	Sediment
Enewetak	E-2	21.6	5-53	22	37	5.8	5.9
Enewetak	E-10	4.4	3-45	14	33	1.3	4.2
Enewetak	E-24	12.0	10-24	20	80	1.5	2.5
Enewetak	E-33	.14	.18-.35	.25	9.1	0.2	0.3
Enewetak	E-37	.24	.04-.21	.11	9.2	0.3	0.1
Enewetak	E-43	1.0	1.1-3.9	2.1	28	0.4	0.8
Ailinginae		.22	.03-.09	.06	0.48	4.6	1.3
Ailuk		.022	.005-.013	.009	0.57	0.4	0.2
Likiep		.013	.003-.011	.007	0.58	0.2	0.1
Rongelap		.49	.12-.53	.28	1.31	3.7	2.1
Rongerik		.059	.03-.17	.086	1.20	0.5	0.7
Wotho		.024	.006-.013	.01	0.42	0.6	0.2

a - $Kd = \frac{pCi/gm\ sediment}{pCi/gm\ water}$.